

## ION PRODUCED COMETARY ORGANIC CRUST

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### ABSTRACT

For several years many experimental results have been obtained on the chemical and physical changes induced by ion and electron irradiation of materials with a view to their Astrophysical relevance (Foti et al., 1984; Johnson et al., 1984; Andronico et al., 1987; Lanzerotti et al., 1987; Strazzulla et al., 1991a; Strazzulla and Johnson, 1991). Among the studied effects, one of particular interest is the formation of an organic refractory residue left over after ion irradiation and warming-up at room temperature. We call this residue IPHAC (Ion Produced Hydrogenated Amorphous Carbon). Although "in situ" infrared spectroscopy points out the formation of new molecular species during bombardment at low temperature (Strazzulla & Baratta, 1991) it is not clear if IPHAC is already formed or if its formation is triggered by temperature increase during warming-up of the irradiated target. Being Raman Spectroscopy a technique particularly suitable for the analysis of carbonaceous materials, we have thought and build-up an experimental apparatus to obtain Raman Spectra of frozen hydrocarbons during ion irradiation (Spinella et al., 1991). The present experimental results point out clearly to the formation of IPHAC already at low T and low energy deposition ( $\approx$  few eV/C-atom).

### EXPERIMENTAL RESULTS

Raman spectroscopy gives valuable information on the effects induced by ions impinging on solids at low temperature. Indeed this technique has both the ability to distinguish between chemical species and can provide valuable evidence to the structural properties of materials and, in particular, of carbonaceous materials (Robertson et al., 1986). Raman spectroscopy has been in fact used to get insight in the structural lattice damage of solids resulting from ion bombardment (Wright et al., 1976).

Details on the experimental set-up we have used have been given elsewhere (Spinella et al. 1991). A scattering chamber was faced through KBr windows, to a Raman SPEX 1488 double monochromator equipped with two holographic gratings (1800 grooves/mm) to which an OMA III intensified reticon or a cooled photomultiplier were faced as detectors. We used a 90 degrees scattering geometry where the direction of incident and collected light, as well as the direction of the ion-beam, are mutually perpendicular. Vacuum was better than  $10^{-7}$  mbar. Frosts were accreted onto a silicon crystal (111) substratum put in thermal contact with a closed-cycle Helium cryostat (10-300 K) by admitting gas into the chamber, through a needle valve. The cold finger has been designed to avoid that the laser (Argon ion gas laser) light, reflected in the specular direction, could be collected by the monochromator collimating lenses. After or during condensation, ices were bombarded by 3 keV  $He^+$  ions. The beam produces a  $2 \times 2$  cm<sup>2</sup> spot on the target. Currents were in the range of few  $\mu$ -ampere/cm<sup>2</sup> or less in order to avoid a macroscopic heating.

In a particular experiment, a benzene ( $C_6H_6$ ) film, (about  $2 \mu m$  thick) obtained by slow rate deposition ( $\approx 0.11 \mu m/min$ ) on the substratum ( $T=77 K$ ), was irradiated with  $3 keV He^+$  ions. Some spectra are reported in Fig. 1: (from top to bottom) as deposited, after  $30 eV/C$ -atom and after warming-up at room temperature. All of the spectra in the figure were obtained by using the  $514 nm$  Argon laser line, with an output power less than  $\approx 40 mW$ . The incident light was with the electric vector orthogonal to the plane of scattering; the scattered light polarization was not analyzed. The entrance slit width was  $0.1 mm$ , corresponding to a resolution of about  $5 cm^{-1}$  and the exposure time was  $3 sec \times 30 scan$  in each spectral region. It is interesting to note that, in the irradiated sample, a newly formed broad band at  $\approx 1600 cm^{-1}$ , typical of amorphous carbon or hydrogenated amorphous carbon (Robertson, 1986; Yoshiwawa et al., 1988), appears in addition to the benzene ones. The appearance of such a structure points-out that IPHAC has been already formed during bombardment at low temperature.

Analogous results have been obtained by irradiating frozen butane ( $C_4H_{10}$ ). A butane film (about  $2 \mu m$  thick), accreted on the silicon substratum ( $T=10 K$ ) at a rate deposition as above, was irradiated with  $3 keV He^+$  ions. The spectra were obtained by using a cooled photomultiplier as detector. Some spectra are reported in Fig. 2: (from top to bottom) as deposited, after  $1.5 eV/C$ -atom and after  $21 eV/C$ -atom; the last spectrum was obtained for the organic residue at  $10 K$ . The slits width was  $0.2 mm$ , corresponding to a resolution of about  $10 cm^{-1}$  and the exposure time was  $0.3 sec$  (with a sampling step of  $2 cm^{-1}$ ). All of the remaining parameter concerning the laser and the scattering geometry were the same as for the benzene experiment. Also in this case the newly formed broad band at  $\approx 1600 cm^{-1}$  appears in addition to the butane ones. The appearance of the structure even in an aliphatic (single bonds) compound like butane, testifies for the generality of the process concerning the formation, at low temperature, of IPHAC, i.e. of a complex mixture of ring molecules linked together by linear chains (Strazzulla & Baratta, 1991).

## CONCLUSIONS

These results may have relevant astrophysical applications, in particular for cometary physics. In fact, the outer layers of a comet, the comet's mantle, is altered by cosmic-ray particle processing of the ices and organics during the comet's  $4.6 \times 10^9$  years residence time in the Oort cloud (Donn, 1976; Wipple, 1977; Strazzulla, 1986; Johnson et al., 1987).

It has been suggested (Johnson et al., 1987; Strazzulla & Johnson, 1991; Strazzulla et al., 1991b) that a comet exposed to background particle radiation in the Oort cloud obtains an outer web of non-volatile material which will lead to the formation of a substantial "crust". The results presented here support the hypothesis that the cometary organic crust can be already formed during the long stay in the Oort cloud and its development does not requires a first passage (heating) in the inner Solar System.

When a new comet enters the inner Solar System there will be early activity, initial fissures in the crust and the break-off of unstable pieces of the crust, due to warming of sub surface species. If this comet enters a periodic orbit in the inner Solar System the remaining mantle should be continuously hardened due to thermal processes (Fanale & Salvail, 1984; Prialnik & Bar-Nun, 1988).

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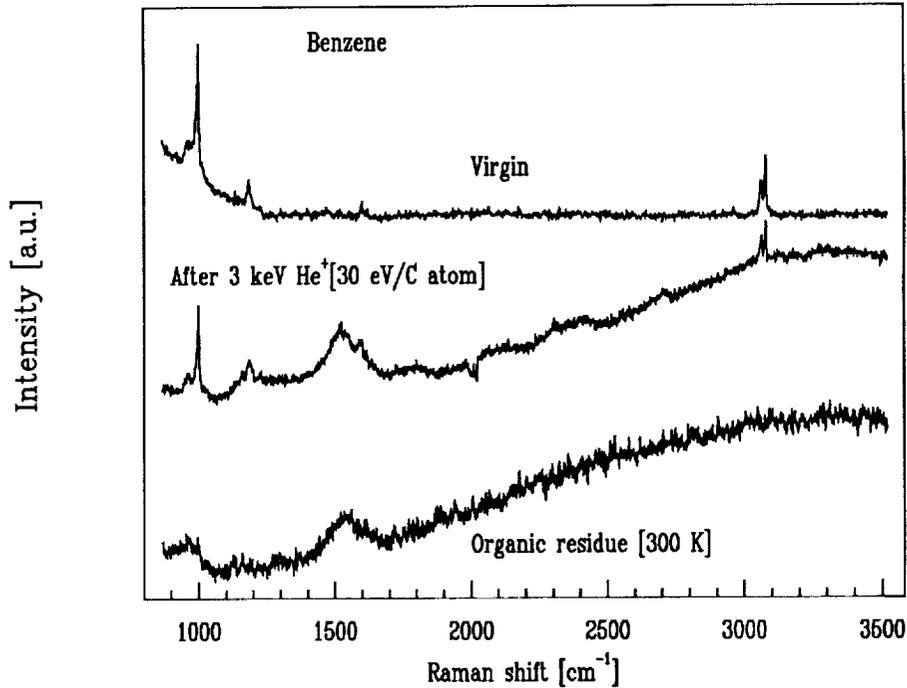


Fig.1 Raman spectra of frozen benzene: as deposited (77 K), during irradiation with 3 keV He ions (77 K) and of the organic residue (300 K) left over after ion irradiation.

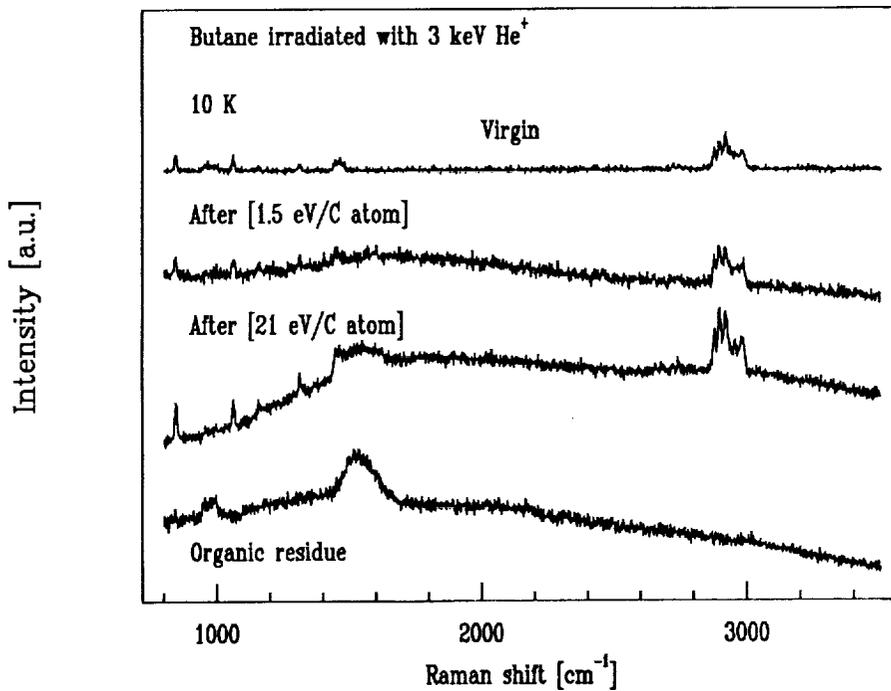


Fig.2 Raman spectra of frozen butane: as deposited (10 K), during irradiation with 3 keV He ions (10 K) and of the organic residue (10 K) left over ion irradiation.

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